The Ripplon-Limited Mobility of Ions Trapped Below the Free Surface of Superfluid Helium

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New measurements are reported of the damping of two-dimensional plasma resonances in circular pools of negative ions trapped below the free surface of superfluid ⁴He at temperatures down to about 15 mK. At the lowest temperatures the damping is determined by a ripplon-limited ionic mobility. The observed dependence of this mobility on temperature and trapping depth is shown to be similar to that found in earlier experiments on pools of positive ions. A theory of the ripplon-limited mobility is described, based on a calculation of the scattering of capillary waves by a single ion. It is shown that the theory accounts for the observed temperature and depth dependences, but that it yields absolute values that are too large by a factor of about 10 for both species of ion. Possible reasons for this discrepancy are discussed, but no satisfactory explanation has been found.

1. INTRODUCTION

This paper is concerned with the damping of two-dimensional plasma modes in circular pools of ions trapped below the free surface of superfluid ⁴He. It follows an earlier paper,[1] which contained a detailed description of the background to this work and of the experimental techniques.

Both positive and negative ions can be trapped below the surface of superfluid ⁴He by a combination of an external electric field, E_0, that forces the ions towards the surface and the image potentials that force them away from it. The ions can be produced conveniently by applying a suitable

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potential to a field emission or field ionization tip immersed in the helium. The positive ion is probably a He_2^+ ion embedded in a small volume of solid helium, formed by electrostriction; it has a radius of about 0.55 nm and an effective mass of about $30 m_4$ (m_4 being the mass of a helium atom); the negative ion is an electron in a bubble; it has a radius of about 1.9 nm and an effective mass of about $237 m_4$. The trapping depth, z_0, is given in terms of the holding field E_0 by

$$z_0^2 = \frac{\Lambda_0}{eE_0}; \qquad \Lambda_0 \approx \frac{(\varepsilon - 1) e^2}{32\pi\varepsilon_0} \qquad (1.1)$$

where ε is the dielectric constant of the helium. We have assumed, as is the case, that $(\varepsilon - 1)$ is small compared with unity. With a set of electrodes that provides a suitable fringing field it is possible to trap a circular pool of ions with a density, n_0, that is almost spatially uniform; typically n_0 is in the range from $3 \times 10^{11} \, m^{-2}$ to $1 \times 10^{12} \, m^{-2}$. Thermally excited vertical motion of the ions in the trapping potential is small compared with the ionic spacing, so that for many purposes the system is two-dimensional. It can therefore support two-dimensional plasma waves in which the ionic motion is horizontal, and in a circular pool of finite extent these waves give rise to standing plasma mode resonances. Detailed studies of these plasma mode resonances were reported in.[1]

The plasma waves can be damped by two mechanisms: that associated with a finite ionic mobility within the helium; and that associated with any internal dissipation, due, for example, to viscosity, within the ionic plasma itself. The damping gives rise to a finite linewidth in the plasma resonant modes. As explained in[1] the two contributions to the damping can be distinguished by their different dependences on plasma frequency, and it was shown that under the experimental conditions used so far the finite ionic mobility is dominant. Measurements of the linewidth can therefore be used to deduce values of the mobility. It was also shown in[1] that at temperatures above about 100 mK the mobility is found to be independent of trapping depth and to be due to the scattering by the ions of the bulk thermal excitations (the phonons and rotons) within the helium. At lower temperatures, however, the mobility is observed to be strongly dependent on trapping depth, which suggests that it becomes limited by the scattering of thermally excited capillary waves, or ripplons, on the surface of the helium. A detailed experimental study of this ripplon-limited mobility, μ_r, for the case of the *positive* ion was reported in.[1] It was shown that the experimental results were consistent with the formula

$$\left(\frac{1}{\mu_r}\right) = \frac{A_+ T^m}{z_0^n} \qquad (1.2)$$

for the ripplon contribution to the inverse mobility, where T is the temperature, $m = 1$, $n = 7/2$ (more precisely $m = 0.99 \pm 0.018$; $n = 3.53 \pm 0.17$), and $A_+ = (6.5 \pm 0.5) \times 10^{27} \, \text{m}^{-3/2} \, \text{V}^{-1} \, \text{s}^{-1} \, \text{K}$. (If the different scattering processes are independent, the contributions to the inverse mobility add.)

In this paper we report corresponding measurements of the ripplon-limited mobility of the *negative* ion. We find that a formula of the form (1.2) is still obeyed, and we report values of the parameters m, n, and A_-; within experimental error the values of m and n are still 1 and 7/2, but A_- is about $55 \times 10^{27} \, \text{m}^{-3/2} \, \text{V}^{-1} \, \text{s}^{-1} \, \text{K}$. We also describe the first stages of the development of a theory of the ripplon-limited mobility. This theory is based on a consideration of the scattering of capillary waves by a single ion; it yields a formula of the form (1.2) with $m = 1$ and $n = 7/2$, but it predicts values of the parameters A_+ and A_- that are too large by factors of order 10. More refined versions of the theory will be published in later papers.

2. THE RIPPLON-LIMITED MOBILITY OF THE NEGATIVE ION: EXPERIMENTAL RESULTS

Measurements of the mobility of the negative ion at low temperatures were obtained from the linewidths of the plasma resonances (or equivalently from the decay time of the freely oscillating plasma modes) as described in.[1] The experimental results are shown in Figs. 1 and 2, where we have plotted the dependence of the mobility on temperature and trapping depth. Fitting the results to a formula of the form (1.2), we find

$$m = 1.26 \pm 0.45; \qquad n = 3.59 \pm 0.10;$$
$$A_- = (55 \pm 5) \times 10^{27} \, \text{m}^{-3/2} \, \text{V}^{-1} \, \text{s}^{-1} \, \text{K} \tag{2.1}$$

Within the experimental error the values of m and n agree with those found for the positive ion. The value of m has been determined with less precision for the negative ion than for the positive because the mobility of the negative ion is ripplon-dominated only for temperatures below about 40 mK, compared with about 100 mK for the positive ion.

3. THEORY OF THE RIPPLON-LIMITED MOBILITY

In this section we give in outline a theory of the ripplon-limited mobility. The theory accounts for the values of m and n, but it gives values of A that are too large by a factor of order ten. The theory is based on a simplified model, but, as we shall explain, the obvious simplifications do not seem to be responsible for its shortcomings. Apart from the use of a

simplified model, our presentation of the theory in this paper will not be fully rigorous. A more detailed and rigorous treatment will be presented in a later paper.

3.1. The Model

The theory relates to the mobility of a single isolated ion, trapped in the potential formed from the holding field E_0 and the image of the ion.
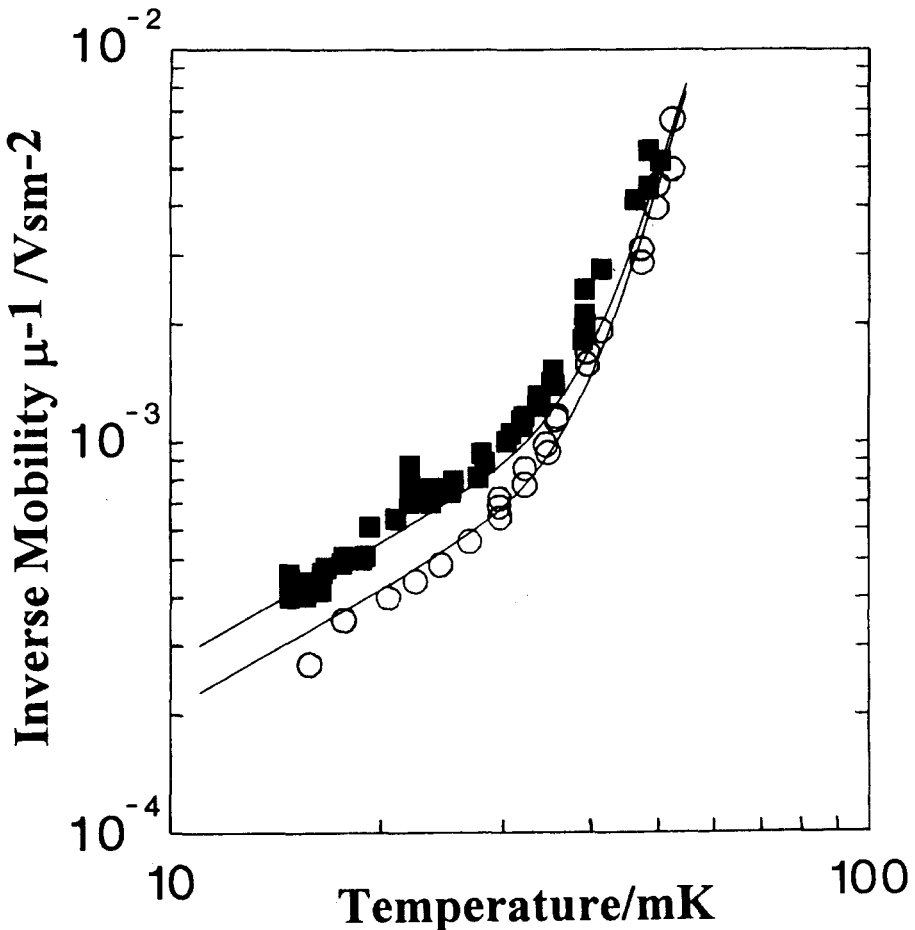

Fig. 1. The inverse mobility of negative ions plotted against temperature for two trapping depths. ○: $z_0 = 18.3$ nm; ■: $z_0 = 16.9$ nm. The solid lines are calculated for a ripplon-limited mobility given by (1.2), with $m = 1$, $n = 7/2$, and $A_- = 5.5 \times 10^{28}$ m$^{-3/2}$ V^{-1} s^{-1}, combined with the known phonon-limited mobility obtained from experiment in.[1]

Vertical distances, z, are measured from the undisturbed surface of the helium. For small vertical displacements from the equilibrium trapping depth $z = -z_0$ (Eq. (1.1)), this potential has the parabolic form

$$V(z) = \text{const} + \frac{eE_0}{z_0}(z + z_0)^2 \qquad (3.1.1)$$

The natural angular frequency of vertical oscillation of the ion in this potential is given by

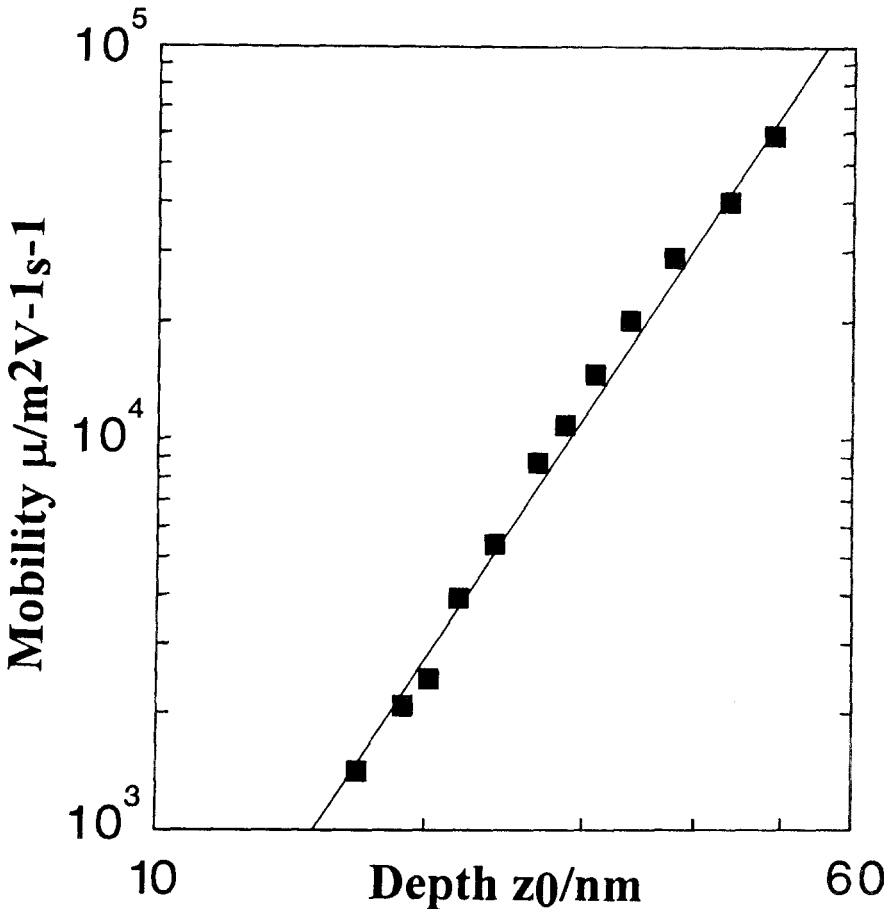$$\omega_0^2 = \frac{2\Lambda_0}{Mz_0^3} \qquad (3.1.2)$$

Fig. 2. The mobility of negative ions plotted against trapping depth for a temperature of 19 mK. The solid line is a plot of (1.2) with the values of m, n, and A_- used in the lines in Fig. 1.

where M is the ionic effective mass. The frequency $\omega_0/2\pi$ is typically 50 MHz. The holding potential, $V(z)$, becomes strongly anharmonic for large vertical displacements, but we shall assume that this is not relevant to our present discussion.

We treat the ripplon scattering as the scattering of classical capillary waves. We assume that a single capillary wave is incident on the ion, and we calculate a scattering cross section. In so doing we assume that the ion remains at rest except for such motion as may be induced by the incident capillary wave. Using this cross section we obtain a mobility by considering the effect of a thermal distribution of ripplons. The ion is assumed to be a rigid sphere of radius b and the helium to be a continuous classical inviscid incompressible fluid of density ρ and surface tension σ. Capillary waves on the surface of this fluid travelling in the x-direction can be described by the velocity potential (Ref. 2, p. 237)

$$\phi = \phi_0 \exp(kz) \exp i(kx - \omega t) \qquad (3.1.3)$$

where ω and k are related by the dispersion relation

$$\omega^2 = \frac{\sigma}{\rho} k^3 \qquad (3.1.4)$$

We have assumed that the effect of gravity can be neglected in (3.1.4), which is justified if $2\pi/k$ is small compared with the capillary length $(\sigma/g\rho)^{1/2}$.

Our neglect of the finite compressibility of the helium can be justified by two comments. First, at the frequencies concerned the ripplon dispersion relation is essentially unaffected by the compressibility of the helium.[8] And secondly, again at the frequencies concerned, for which the wavelength of sound is much larger than the ionic radius, the flow round the ion is also to a very good approximation unaffected by this compressibility (Ref. 2, p. 280).

3.2. The Scattering Mechanisms

Capillary waves are scattered by an ion through a combination of two effects, one *hydrodynamic* and one *electrostatic*.

In the presence of a capillary wave there is generally an oscillating relative motion between the ion and the fluid in its immediate neighbourhood. A boundary condition at the surface of the ion requires that there be no normal component of this relative velocity. Provided that, as is the case, the relevant capillary waves have wavelengths large com-

pared with the ionic radius, the boundary condition can be satisfied if there is generated around the ion an appropriate oscillating dipolar velocity field. This oscillating dipolar velocity field generates the *hydrodynamic* contribution to the scattered capillary wave.

The *electrostatic* effect arises from the interaction between the charged ion and the polarization charge on the free surface of the helium. Motion of the surface in the capillary wave gives rise to an oscillating electrostatic force on the ion, and the resulting oscillatory motion of the ion gives rise to an oscillating force on the surface of the helium and hence to a scattered wave.

3.3. A Simplified Treatment

A full and rigorous treatment of these effects will be published in a separate paper. It will be shown that both effects make important contributions to the scattering, but that the mobility is limited almost entirely by ripplon scattering at frequencies close to the resonant frequency ω_0. In this resonant scattering process the velocity of the ion relative to the fluid is due almost entirely to the resonant vertical motion of the ion, so that both the fluid velocity and the horizontal motion of the ion can be neglected in the calculation of the hydrodynamic contribution to the re-radiated capillary wave.

This being the case, we can obtain the scattering cross section in a greatly simplified way, which we now describe.

A capillary wave of the form (3.1.3) is incident on the ion, and we assume that the wave is only weakly perturbed by the ion. We calculate the induced motion of the ion when ω is close to ω_0 (and $k = k_0$). The motion of the ion will be damped, by radiation of capillary waves (and possibly by other processes), and we assume that this damping can be described by a relaxation time τ_z. The detailed calculations show that the radiation damping is small, so that $\omega_0 \tau_z \gg 1$. We can then calculate the rate at which energy is being extracted from the capillary wave, and hence we obtain an effective cross section, σ_E, relating to this extraction of energy, which is given by

$$\sigma_E = \frac{2Mk_0^2 \omega_0 \tau_z}{3\rho} \cdot H_z^2(k_0 z_0) \cdot \frac{1}{1 + 4(\omega - \omega_0)^2 \tau_z^2} \qquad (3.3.1)$$

where

$$H_z(\gamma) = \frac{2\pi\rho b^3}{M} \exp(-\gamma) - \frac{\rho \Lambda_0}{M\sigma\gamma} K_2(\gamma) \qquad (3.3.2)$$

and K_2 is the modified Bessel function.[3] The detailed derivation of this equation is given in an appendix.

The ionic mobility can be obtained from a transport cross section σ_T, with the formula

$$\frac{e}{\mu} = -\frac{\hbar}{4\pi} \int k^3 \frac{\partial n_k}{\partial \omega} v_G \sigma_T \, dk \qquad (3.3.3)$$

where $v_G = d\omega/dk$ is the ripplon group velocity, and n_k is the ripplon distribution function at temperature T, given by

$$n_k = \left[\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1 \right]^{-1} \qquad (3.3.4)$$

Equation (3.3.3) is the two-dimensional version of an equation derived by Baym *et al.*[4] We assume that the two cross sections, σ_E and σ_T are equal, an assumption that is valid if the capillary waves are scattered isotropically, which is indeed the case. Substituting from (3.3.1) into (3.3.3), and using the fact that the integrand in (3.3.3) is very small unless $\omega \approx \omega_0$ and $k \approx k_0$, we find that

$$\frac{e}{\mu} = \frac{Mk_0^{7/2}k_B T}{12\rho^{1/2}\sigma^{1/2}} \cdot H_z^2(k_0 z_0) \qquad (3.3.5)$$

the relaxation time τ_z having disappeared in the integration. The more rigorous treatment of this model gives the same result.

4. DISCUSSION AND COMPARISON WITH EXPERIMENT

We see from Eqs. (1.1), (3.1.2), and (3.1.4) that the product $k_0 z_0$ is independent of the trapping depth z_0. It follows from Eq. (3.3.5) that μ should be proportional to $z_0^{7/2}$, in agreement with experiment. Equation (3.3.5) leads also to the prediction that the inverse mobility is proportional to the temperature, again in agreement with experiment. However, the predicted values of A_+ and A_- are $122 \times 10^{27} \text{ m}^{-3/2} \text{ V}^{-1} \text{ s}^{-1} \text{ K}$ and $316 \times 10^{27} \text{ m}^{-3/2} \text{ V}^{-1} \text{ s}^{-1} \text{ K}$ respectively; which are larger than the experimental values by factors of about 19 and 6 respectively.

The dependence of mobility on trapping depth arises in the theory in a quite subtle way, and it seems unlikely that the agreement here with experiment can be an accident. This suggests strongly that the theory incorporates much of the correct basic underlying physics. In particular, the idea that the dominant ripplon scattering is at the resonant frequency ω_0 seems sound. Nevertheless, the failure of the theory to account correctly for the absolute magnitude of the mobility shows that some error has crept in.

The theory is based on a number of simplifying assumptions. We examine whether these assumptions are likely to have had a serious effect on our final result.

4.1. The Effect of Ionic Interactions and Ionic Thermal Motion

The theory deals only with a single isolated ion, which is at rest except for the motion induced by the incident capillary wave. In reality there is an array of ions, which is usually in a crystalline phase at the temperatures concerned,[5] and the ions are undergoing thermal motion.

The ions are coupled together through Coulomb forces. In the crystalline phase the motions of the ions must therefore be described in terms of normal modes. The normal modes for motion in the horizontal plane are the longitudinal plasma modes together with transverse shear modes.[6] The normal modes for vertical motion depend on the extent to which the holding field E_0 is uniform.[6] If each ion is exposed to exactly the same holding field, there is a band of wave-like modes, with a band width of about 2 MHz ([6] and Ewbank, private communication), the frequency being equal to ω_0 only in the limit of zero wavenumber. If the holding field is not exactly uniform the situation is more complicated, and the normal modes associated with vertical ionic motion may become localized. At a finite temperature all the normal modes will be thermally excited. A strictly correct theory ought therefore to consider the scattering of ripplons by the whole array of coupled ions, with the additional complication that the ions are moving in a way that corresponds to thermal excitation of the normal modes of the system. Believing, as we have explained, that the scattering responsible for the ripplon-limited mobility must be closely associated with the resonant response of the ions at the frequency ω_0, we shall confine our attention to processes that involve the (resonant) absorption or emission of the quanta that are associated with the modes of vertical oscillation. We shall denote such a quantum by the letter V. We denote a ripplon by the letter R, and we denote a plasmon or transverse phonon (with motion in the plane of the ion-pool) by the letter P.

We shall assume that the holding field E_0 is uniform, so that the modes of vertical oscillation are characterized by a wave vector $\mathbf{k}_V$. As we have explained, the frequency ω_0 becomes dependent on $\mathbf{k}_V$, and we denote it by $\omega_V(\mathbf{k}_V)$, where $\omega_V(0) = \omega_0$.

We shall suppose first that the plasmons and transverse phonons are not involved, which might well be the case if the crystal is at zero temperature. The simplest scattering process can then be written

$$R \rightarrow V + G \rightarrow R' + G' \qquad (4.1.1)$$

and this will be associated with the intersection of the dispersion curves $\omega_R(\mathbf{k}_R)$ and $\omega_V(\mathbf{k}_V + \mathbf{G})$, where $\mathbf{G}$ is a reciprocal lattice vector. However, the effect of this level crossing is simply to modify the *shapes* of the two dispersion curves through hybridization, in the manner usual at level crossings. We note in passing that the ripplon dispersion curve is also modified if $\mathbf{k}_R$ lies on or near a Brillouin zone boundary of the crystal. These modifications to the dispersion curves do not lead to any scattering that will limit the ionic mobility.

Higher order scattering processes involving three or more ripplons, such as

$$R + R' \rightarrow V + G \rightarrow R'' + G' \tag{4.1.2}$$

do limit the mobility, but they rely on a non-linear ripplon-ripplon coupling and would lead to an inverse mobility that is not simply proportional to the temperature and therefore not in agreement with experiment.

We turn therefore to processes that involve plasmons and transverse phonons. We note in passing that ripplon scattering by an isolated ion that is in motion will lead to a Doppler shift in the ripplon frequency (although the shift is small compared with ω_0), so we might expect that scattering from a ion crystal in which there is thermal motion of the ions will also lead to inelastic scattering, with the emission or absorption of plasmons or transverse phonons. Therefore we consider processes of the type

$$R + P_1 + P_2 + \cdots \rightarrow V + G + P_3 + P_4 \cdots \rightarrow R + G' + P_5 + P_6 + \cdots \tag{4.1.3}$$

In the scattering of waves (in this case capillary waves) from a crystal the extent to which processes of this type take place is determined by the Debye-Waller factor $\exp(-2W)$; $2W$ is given by

$$2W = \tfrac{1}{3}K^2 \langle u^2 \rangle \tag{4.1.4}$$

where K is the magnitude of the scattering vector and $\langle u^2 \rangle$ is the mean square atomic displacement. For values of K corresponding to the scattering of ripplons of frequency ω_0 through a large angle, and for the temperatures used in the experiments described in this paper, the Debye-Waller factor for the ion crystal turns out to be very small (K is larger by a factor of about 5 than the magnitude of the smallest reciprocal lattice vector). This means that there is practically no scattering into the Bragg peaks; almost all the scattering is inelastic and involves the absorption or emission of one or more phonons. It follows that processes of the type (4.1.3) must be dominant.

The Debye-Waller factor is small because the right hand side of (4.1.4) is large. This means that the difference in phase between capillary waves scattered from adjacent ions fluctuates more or less randomly over the crystal by an amount large compared with unity. It follows that the total scattering of the capillary waves through large angles is practically the same as one would find for an assembly of ions that are positioned at random: the scattered intensity is that due to a single ion multiplied by the number of ions. Strictly speaking the single-ion scattering should take account of the fact that the ions are in horizontal motion, which leads, as we have noted, to a Doppler shift in the frequency of the scattered capillary waves, but it can be shown that the effect on the scattering cross-section is small. Thus we conclude that a calculation in which we treat the ions as independent scatterers ought to be valid, and that the result (3.3.5) ought to be unaffected by the presence of any spatial order in the ionic array or of any thermal motion of the ions.

Experimental evidence that this is indeed the case is provided by the experimental observation that the ripplon contribution to the inverse mobility remains proportional to the temperature even through the melting temperature; there is no anomaly in the ripplon-limited mobility at the melting temperature of the crystal.

4.2. The Effect of Anharmonicity in the Holding Potential

The vertical holding potential is approximately harmonic only for small vertical displacements of the ions, significant departures from the harmonic form occurring at displacements that can be thermally excited even at the low temperatures relevant to the present problem. However, any large effect of such anharmonicity would be expected to increase rapidly with increasing temperature and so destroy the predicted proportionality between inverse mobility and temperature.

4.3. The Effect of Finite Ripplon Lifetimes

The theory takes no account of any finite ripplon lifetime, arising from ripplon-ripplon interactions. However, theory indicates that this lifetime is strongly temperature dependent,[7] so that any effect of a finite ripplon lifetime on the mobility would also be strongly temperature dependent and would therefore again destroy the predicted proportionality between inverse mobility and temperature.

We conclude that the simplifications introduced in the theory are unlikely to have led in any obvious way to a significant error in the predicted mobility, without at the same time destroying the agreement with the

observed dependences on temperature and trapping depth. It seems likely therefore that some subtle effect is at work, but we have failed so far to discover what it is.

APPENDIX

We describe the calculations that lead to Eq. (3.3.1).

We use cylindrical polar coordinates (r, θ, z), in which the positive z-axis points vertically upwards, the undisturbed surface of the helium is at $z = 0$, and the ion in equilibrium is at $r = x_0$, $\theta = 0$, $z = -z_0$. The incident capillary wave travelling in the direction $\theta = 0$ is described by the velocity potential

$$\phi_i = \phi_0 \exp(ikr \cos \theta - i\omega t) \exp(kz) \tag{A.1}$$

(cf. Eq. (3.1.3)), and the corresponding surface displacement in the z direction is given by

$$\zeta = \zeta_0 \exp(ikr \cos \theta - i\omega t) \tag{A.2}$$

where

$$\zeta_0 = \frac{ik}{\omega} \phi_0 \tag{A.3}$$

Motion of the ion is induced by the two effects described in Sec. 3.2, which we call the *hydrodynamic* and *electrostatic* effects. The motion is in the plane $\theta = 0$, and we denote the displaced position of the ion by the coordinates $r = x_1$, $\theta = 0$, $z = -z_1$.

A.1. The Hydrodynamic Effect

Let the velocity field derived from the potential (A.1) be $\mathbf{v}$ (i.e., $\mathbf{v} = \nabla \phi_i$). This is the field that exists in the absence of the perturbation due to the ion. The equation of motion of the ion is then (Ref. 2, p. 35)

$$M \frac{d\mathbf{V}}{dt} = 2\pi \rho b^3 \left(\frac{\partial \mathbf{v}}{\partial t} \right)_{\text{ion}} + \mathbf{f} \tag{A.4}$$

where $\mathbf{V}$ is the velocity of the ion, the time derivative of the fluid velocity is evaluated at the position of the ion, and $\mathbf{f}$ is any external force applied to the ion. For a model of the ion in which the bare ion is a hard sphere, the surrounding superfluid is incompressible, and the normal fluid density is very small, the effective mass M is the sum of the bare mass and half the mass of fluid displaced. In practice a value of M can be obtained from experiment.[1]

A.2. The Electrostatic Effect

In order to obtain the force on the ion due to electrostatic forces we first evaluate the force on the liquid due to the electric field of the ion, and then use the fact that this must be equal and opposite to the electrostatic force on the ion.

We calculate from first principles the electrostatic force on the helium due to an externally applied electric field $\mathbf{E}_0$, and we introduce some clearly defined approximations. Using the fact that a polarized dielectric (polarization $\mathbf{P}$) is equivalent to a charge density equal to $-\operatorname{div}\mathbf{P}$ throughout its volume together with a charge per unit area over its surface equal to the normal component of $\mathbf{P}$, we find that the total force on the helium can be written

$$\mathbf{F} = \int \mathbf{E}_0(\mathbf{P} \cdot d\mathbf{S}) - \int \mathbf{E}_0 \operatorname{div} \mathbf{P} \, d\tau \tag{A.5}$$

where the two integrals are over the surface and the volume of the helium respectively. With the use of some straightforward vector identities, and the fact that $\operatorname{curl} \mathbf{E}_0 = 0$, this is easily transformed to

$$\mathbf{F} = \int (\mathbf{P} \cdot \nabla) \mathbf{E}_0 \, d\tau \tag{A.6}$$

For a dielectric that is described by a dielectric constant ε, $\mathbf{P} = \varepsilon_0(\varepsilon - 1) \mathbf{E}$, where $\mathbf{E}$ is the total electric field in the material, due to both $\mathbf{E}_0$ and the field produced by the polarization. Hence we have

$$\mathbf{F} = \varepsilon_0(\varepsilon - 1) \int (\mathbf{E} \cdot \nabla) \mathbf{E}_0 \, d\tau \tag{A.7}$$

But $\mathbf{E}$ differs from $\mathbf{E}_0$ by terms proportional to $\mathbf{P}$ and hence to the susceptibility $(\varepsilon - 1)$. For liquid helium $(\varepsilon - 1)$ is equal to 0.0572, so if we are con-

tent with results that are accurate to within a few percent we can neglect terms of order $(\varepsilon - 1)^2$. Equation (A.1.4) then reduces to

$$\mathbf{F} = \tfrac{1}{2}\varepsilon_0(\varepsilon - 1) \int \nabla(\mathbf{E}_0^2) \, d\tau = \tfrac{1}{2}\varepsilon_0(\varepsilon - 1) \int \mathbf{E}_0^2 \, d\mathbf{S} \qquad (A.8)$$

where we have again used curl $\mathbf{E}_0 = 0$.

Using Eq. (A.8) we find that the change in electrostatic potential energy when a charge e is brought from $r = 0$, $z = -\infty$ to $r = 0$, $z = -z_1$ in the presence of a surface displacement $\zeta(r, \theta)$ (A.2) is given by

$$U = \frac{e^2(\varepsilon - 1)}{32\pi^2\varepsilon_0} \int_{-\infty}^{-z_1} dz \int_0^\infty d^2r \, \frac{1}{[r^2 + (|z| + \zeta)^2]^2} \qquad (A.9)$$

Therefore the change in potential energy if ζ is small is given by

$$\delta U = -\frac{e^2(\varepsilon - 1)}{32\pi^2\varepsilon_0} \int_\infty^{z_1} dz \int_0^\infty d^2r \, \frac{\partial}{\partial z}\left[\frac{1}{(r^2 + z^2)^2}\right] \zeta(r, \theta) \qquad (A.10)$$

$$= -\frac{e^2(\varepsilon - 1)}{32\pi^2\varepsilon_0} \int_0^\infty d^2r \, \frac{1}{(r^2 + z_1^2)^2} \zeta(r, \theta) \qquad (A.11)$$

If $\zeta(r)$ is given by (A.2) the integral in (A.11) can be easily evaluated in terms of Bessel functions. Making the simple generalization required if the ion is at $r = x_1$ instead of $r = 0$, we obtain

$$\delta U = -\frac{\Lambda_0 k}{z_1} K_1(kz_1) \, \zeta_0 \exp i(kx_1 - \omega t) \qquad (A.12)$$

where K_1 is a modified Bessel function.[3]

It follows that the components of the electrostatic force acting on the ion in the presence of the surface displacement (A.2) are given by

$$F_x = -\frac{\partial \delta U}{\partial x_1} = \frac{i\Lambda_0 k^2}{z_1} K_1(kz_1) \, \zeta_0 \exp i(kx_1 - \omega t) \qquad (A.13)$$

and

$$F_z = \frac{\partial \delta U}{\partial z_1} = \frac{\Lambda_0 k^2}{z_1} K_2(kz_1) \, \zeta_0 \exp i(kx_1 - \omega t) \qquad (A.14)$$

A.3. The Equation of Motion of the Ion

The overall equation of motion of the ion is obtained from Eq. (A.4) by substituting in place of **f** the sum of the force given by (A.13) and (A.14)

and the force derived from the static holding potential (3.1.1). Writing the components of the ionic displacement parallel and perpendicular to the surface as X and Z respectively, and assuming that these displacements are small, we find

$$\ddot{X} + \frac{1}{\tau_x}\dot{X} = \mathbf{k}\omega \cdot H_x(kz_0)\,\phi_0\exp i(kx_0 - \omega t) \qquad (A.15)$$

and

$$\ddot{Z} + \frac{1}{\tau_z}\dot{Z} + \omega_0^2 Z = -ik\omega \cdot H_z(kz_0)\cdot\phi_0\exp i(kx_0 - \omega t) \qquad (A.16)$$

where

$$H_x(\gamma) = \frac{2\pi\rho b^3}{M}\exp(-\gamma) - \frac{\rho\Lambda_0}{M\sigma\gamma}K_1(\gamma) \qquad (A.17)$$

and $H_z(\gamma)$ is given by Eq. (3.3.2). The relaxation times τ_x and τ_z describe the damping of ionic motion in the horizontal and vertical directions respectively. We have made use of the fact that ω and k are related by the dispersion relation (3.1.4). As will be proved in detail in a later paper, ionic motion in the horizontal direction contributes little to the ripplon-limited inverse mobility, and we need not therefore make further use here of Eq. (A.15).

A.4. Calculation of the Cross Section σ_E

It is easily proved that the total energy per unit area in the capillary wave described by Eq. (A.1) is equal to $(1/2)\,\rho k\phi_0^2$ (half the energy is kinetic and half is due to increased surface area). It follows that the energy flux (per unit length) associated with the capillary wave is given by

$$W = \frac{1}{2}\rho k\phi_0^2\frac{d\omega}{dk} = \frac{3}{4}\rho\omega\phi_0^2 \qquad (A.18)$$

$d\omega/dk$ being the group velocity of the wave. At the same time the rate at which energy is being absorbed by the ion is given by

$$\frac{dE}{dt} = \frac{M}{2\tau_z}\cdot|\dot{Z}|^2 \qquad (A.19)$$

Using Eq. (A.16), we find that

$$\frac{dE}{dt} = \frac{k^2\omega^4 M}{2\tau_z}\cdot H_z^2(kz_0)\cdot\phi_0^2\cdot\frac{1}{(\omega_0^2 - \omega^2)^2 + \dfrac{\omega^2}{\tau_z^2}} \qquad (A.20)$$

It follows that the cross-section σ_E is given by

$$\sigma_E = \frac{2Mk^2\omega^3}{3\rho\tau_z} \cdot H_z^2(kz_0) \cdot \frac{1}{(\omega_0^2 - \omega^2)^2 + \omega^2/\tau_z^2} \tag{A.21}$$

Equation (3.3.1) follows immediately if $\omega_0\tau \gg 1$ and $\omega \approx \omega_0$.

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